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THE ROLE OF RECIPROCITY IN THERMAL RADIATION DAMAGE


<u>Problem</u>: "Reciprocity" in thermal radiation studies implies a critical exposure level which is independent of irradiance, using an arbitrary criterion to establish this critical level. The purpose of this work was to point out similarities in behavior of a few diverse materials, to aid in an understanding of this behavior in terms of theoretical solutions, and to indicate the usefulness of the empirical approach.

<u>Method</u>: The origin of the reciprocity condition in experimental studies is briefly examined and its significance in thermal radiation studies is considered. Theoretical calculations on semi-infinite and insulated-slab heat-flow models in one dimension are presented as a guide for later considerations. Experimental data are presented which were obtained by exposing five different types of material to radiation from the carbon-arc and solar furnace imaging sources. These data are then analyzed for their reciprocity "failures".

<u>Conclusions</u>: The method of thermal data presentation which plots the log of critical exposure level against log of pulse length is general enough for data comparison and some behavior identification. Also, the use of a theoretical criterion such as achievement of an arbitrary maximum temperature yields valuable insight into reciprocity deviations. Most materials cannot usually be expected to show reciprocity over the normal range of pulses used in thermal radiation studies. An application of the empirical approach which would make it possible to create useful thermal simulators can be based on the "reciprocity" behavior, and the success of simulation can be measured by similar curves of critical exposure versus pulse duration.

# THE ROLE OF RECIPROCITY IN THERMAL RADIATION DAMAGE

## I. INTRODUCTION

The concept of reciprocity occurs throughout the literature of radiation effects, regardless of the spectral region involved or the nature of the radiation. Reciprocity is usually assumed to imply a critical radiation exposure level which is independent of radiation intensity, using a suitably defined criterion to establish this critical level. Departure from such an independent relationship is often termed "reciprocity failure", and much of the work pertaining to radiation effects or radiation damage is concerned with detailed investigations of reciprocity failure.

This paper deals with a comparison of various damaging effects produced by radiation in the visible and near infra-red regions of the electromagnetic radiation spectrum, usually referred to as "thermal radiation", which has its origin in luminous sources at effective temperatures near 6000°K. The criteria of damage are different for each material, but are generally classified according to the appearance of specific discernible phenomena at or near the surface of exposed samples. In addition, idealized materials and effects are discussed in order to gain perspective in understanding the mechanisms of thermal radiation damage.

## II. OTHER RECIPROCITY INVESTIGATIONS

### A. Photographic Emulsions

The failure of photographic emulsions to show a constant sensitivity at all intensities of incident light led early investigators to study the reciprocity problem. Although the exposure level concerned is much

less than that encountered in thermal radiation damage, it is nevertheless useful to examine the behavior of a typical emulsion and the reasons for its reciprocity failure.

The formation of the photographic effect, measured by the density of the developed emulsion, depends upon a photochemical reaction with the incident radiation, whereby several quanta of radiation are absorbed by a silver halide grain, rendering it developable. The latent image consists of free silver atoms which congregate at sensitized spots on the grains due to the excess of electronic charge produced in the reaction of quanta with the halide. During development the exposed silver halide grains are reduced to metallic silver preferentially, leading to an optical density which increases with exposure and is proportional to the number of silver grains per unit volume.

Although the amount of the product in a photochemical reaction depends only upon the absorbed energy and not on the rate of absorption, it is found that the developed density of a photographic emulsion does not follow such a reciprocity relationship. The data in Fig. 1 show the variation in exposure level required for constant density over a range of exposure durations at .546 microns. (This form of data presentation is one which we shall use also for thermal damage.) These data were taken from curves reproduced by James and Higgins[1] and plotted originally by Webb[2]. The criterion for the exposure levels determined at each exposure time was that the same (medium) density be measured. This particular emulsion shows a minimum in the critical exposure level at about 3 seconds; the extensive data accumulated by Webb showed that the form of this curve was quite dependent upon

temperature and wave length. However, we shall not discuss these variations here.

The accepted explanations of reciprocity-law failure in this case apply separately to the long exposures (low-intensity) and short exposures (high-intensity). The former is believed to be caused by thermal disintegration of the latent image through ejection of electrons at the sensitized regions, with subsequent diffusion of silver ions. An alternative explanation attributes this low-intensity failure to recombination of the silver and halide ions. In either case, the length of the exposure and the availability of some thermal energy are necessary.

The high-intensity reciprocity failure is apparently caused by sluggishness of the migrating silver ions, which cannot keep up with the rate of liberation of electrons and thus limits the rate of growth of sensitized areas on the grain surface. Thus both the high- and low-intensity regions can be qualitatively explained in terms of the photochemical mechanism which governs the fundamental process. Hopefully, we shall try to use similar approaches to the thermal radiation problems, although photochemical effects probably do not play an important role.

B. Skin Erythema Produced by UV-Radiation

Claesson, Juhlin and Wettermark [3,4] investigated the critical dosage required to produce erythema in living mouse skin over a very wide range of intensities, using ultra-violet light from a flash photolysis source and also from a high-pressure mercury arc. The criterion in these exposures was the leakage into the irradiated areas of intravenously-injected Evans blue, which was graded and evaluated independently by four persons. Such

leakage occurs because of capillary damage, and is directly related to the degree of erythema. The minimum exposure which caused blueing was measured for each source and at each intensity, and it was found that this critical exposure remained constant at about .01 cal $cm^{-2}$ over an intensity range of 7 orders of magnitude. The exposure times used varied from 50 microseconds to 700 seconds.

The measurement of a constant critical exposure level over such a large range apparently suggests that the reciprocity relationship is obeyed. Two factors are significant in this conclusion, however. The first is that the wavelength region is restricted to the ultra-violet. Most of the data were taken with filters in the source beam in order to restrict the effective radiation to a region from 250 to 360 millimicrons, for both sources. The maximum intensity for this interval was taken to occur at 310 millimicrons. Studies made by Blum[5] and in this laboratory by Cotton, Gray and Penniman[6] indicate that ultra-violet light produces specific photochemical reactions within the epidermis, and that little of the erythema so induced is caused by heating of the skin. This would tend to produce agreement with the reciprocity law.

The second factor to consider is that no data were taken in the region of pulse durations from 150 microseconds up to about 10 seconds. As we shall see, this covers the region of interest in thermal irradiation studies, where the principal effect on all materials is not photochemical, but is caused by absorption and subsequent heating. Thus even if heating did play a role in UV absorption, the pulse times used would not correspond to those where the reciprocity failure is most critical.

## III. THERMAL RADIATION DAMAGE

### A. Effects of Thermal Irradiation

The response of a material to incident thermal radiation may vary from a slight change in surface appearance and structure to complete destruction of the material or sustained ignition. In order to be able to make quantitative measurements of material response it is necessary to classify these responses according to their nature or to the mechanisms which produce them, and it is also necessary to delineate objective criteria so that the minimum exposures required for their production can be determined. Usually a criterion is sought which is easily recognized, either visually or through an auxiliary measurement, and which is sufficiently important in the degradation process to give a significant degree of change. All of the changes employed herein are irreversible, although it is certainly possible to utilize reversible phenomena. No general statements can be made regarding the rate at which the various phenomena proceed, but reversible processes can probably be described in terms of reactions of various orders.

In most cases of thermal radiation damage we attempt to determine the exposure level at which the particular criterion selected just begins to occur. This corresponds to the concept of a threshold exposure, even though the accuracy of determination of such thresholds may be quite low due to poor experimental conditions or imprecise identification of the criterion. In some cases, however, such as skin burns, a threshold exposure is not even defined since the occurrence of burns is believed to be governed by a statistical distribution of susceptibilities. In these cases the form of analysis used leads to an effective exposure level at which a certain fraction

of the exposed samples will respond to the radiation according to the selected criterion. This type of analysis, commonly used in biological assay studies[7,8], could actually be applied to any of the threshold experiments having experimental errors from various causes, but for data in which the criterion appears suddenly, it is probably superfluous.

The materials in which we are principally interested react to thermal radiation as a direct result of the absorption of radiant energy and the subsequent increase in temperature of the medium. Most of the materials are opaque to the radiation frequencies involved so that the absorption takes place at or very near the front surface of the medium. Some effects of diathermancy (partial transparency with scattering) do exist, but these will not be of primary concern in this study. Likewise, for the very short wavelengths encountered in the radiation from typical sources, there may be some photochemical reactions which proceed independently of the temperature history, but again these will not be of primary concern. Selective absorption of the radiation certainly takes place for most of the materials, but we shall assume that radiation not absorbed is simply reflected and that an absorption coefficient could be measured which would be constant throughout the measurements.

## B. Temperature Thresholds in Ideal Media

Although none of the materials which in practice are exposed to damaging levels of thermal radiation behave like simple physical heat conductors, it is nevertheless useful to calculate the properties of such ideal media in which no change of state or irreversible change takes place. Since our only interest in such media is in the examination of a reciprocity

relation for some arbitrarily defined criterion, we shall determine the exposure $Q_p$ required to produce a given maximum temperature rise $w_m$ during a rectangular pulse with duration $t_p$. This temperature may occur at a particular location x, either in a one-dimensional semi-infinite solid or in a finite slab of thickness L, also one-dimensional.

The opaque, homogeneous, semi-infinite medium which is irradiated at a constant irradiance H, has a temperature rise given[9] by

$$w = \frac{2aH}{k} \sqrt{\alpha t} \ \mathrm{ierfc} \ \frac{x}{2\sqrt{\alpha t}} \tag{1}$$

where a is the absorption coefficient, $\alpha$ is the thermal diffusivity, t is the time, k is the thermal conductivity, and x is the distance measured from the front surface. When the pulse is cut off at $t = t_p$ it has been demonstrated[10] that the temperature after $t_p$ can be represented by

$$w = \frac{2aH}{k} \left[ \sqrt{\alpha t} \ \mathrm{ierfc} \ \frac{x}{2\sqrt{\alpha t}} - \sqrt{\alpha(t-t_p)} \ \mathrm{ierfc} \ \frac{x}{2\sqrt{\alpha(t-t_p)}} \right] \tag{2}$$

The maximum temperature is attained following the end of the pulse, at a time which varies both with x and $t_p$, the pulse length. For the front surface, x = 0, the maximum temperature occurs at $t_p$, so that

$$w_m = \frac{2aH}{k} \sqrt{\alpha \, t_p} \quad (.564) \tag{3}$$

Thus the exposure to attain $w_m$ at x = 0 is given by

$$\begin{aligned} Q_p &= Ht_p \\ &= \frac{w_m \, k}{1.128 \, a\sqrt{\alpha}} \sqrt{t_p} \end{aligned} \tag{4}$$

At a depth x beneath the surface, the maximum temperature is attained at some time later than $t_p$, which we shall designate as $t_m$. This can be found by differentiating (2) with respect to time and setting equal to zero. However, it was found easier in practice to evaluate the points near $w_m$ numerically and thus determine values of $t_m$ and $w_m$ for several values of $t_p$ and $\alpha$ within our range of interest. Under these circumstances the critical exposure is given by

$$Q_p = \frac{w_m k}{2a\sqrt{\alpha}} \quad \frac{t_p}{\left[\sqrt{t_m}\ \mathrm{ierfc}\ \frac{x}{2\sqrt{\alpha t_m}} - \sqrt{t_m - t_p}\ \mathrm{ierfc}\ \frac{x}{2\sqrt{\alpha(t_m - t_p)}}\right]} \tag{5}$$

Equations (4) and (5) are plotted in Fig. 2 for two cases, showing the typical behavior of these idealized media. Curves S-I(1) and S-I(2) show the front surface, x = 0, and sub-surface depth, x = .05 cm, respectively, for $w_m = 500\,^\circ C$, $\alpha = .001\ cm^2\ sec^{-1}$, $k = .001\ cal\ cm^{-1}\ sec^{-1}\ {}^\circ C^{-1}$, and a = 1. Neither of these curves shows a reciprocity obedience, but the curve for a sub-surface position goes through a minimum which is quite flat over a single order of magnitude in pulse duration. At short pulse lengths this critical exposure begins to increase, indicating that larger exposures would be required to produce temperature-dependent effects beneath the surface as the time of delivery was shortened significantly. At long pulse lengths the critical exposure at a sub-surface depth approaches that required at the surface, in agreement with the existence of a smaller temperature gradient within the medium. The occurrence of the minimum in the $Q_p$ vs. $t_p$ curve is a function of the parameter

$$S = \frac{x}{2\sqrt{\alpha}} \tag{6}$$

which has the value 0.79 in this particular case. Changes in $w_m$ and the absorptance simply shift the curves along the $Q_p$ axis. Changes in $\alpha$ shift curve S-I(1) similarly, with the slope on the log $Q_p$ vs. log $t_p$ curve remaining constant.

The larger separation in $Q_p$ of these two curves for short pulses indicates that any phenomenon which is temperature dependent must be very sensitive to the effective value of S existing for the material, and that appropriate obedience to the reciprocity law can be attained by phenomena which occur at a threshold temperature beneath the surface. A temperature-dependent effect which occurs <u>only</u> on the surface would not be expected to show reciprocity at all, unless it had a separate time-dependent mechanism.

Most actual samples encountered in studies of thermal radiation damage are not extensive in total thickness, so that the use of the above expressions for a semi-infinite medium may not be sufficiently accurate. We must therefore examine the similar relations for a finite, opaque, homogeneous slab of thickness L, assuming that it is insulated from its surroundings and irradiated at the face x = 0 by a beam of flux density H. Carslaw and Jaeger[9] give for the temperature rise at x in such a slab

$$w = \frac{2aH}{k} \sqrt{\alpha t} \sum_{n=0}^{\infty} \left\{ \text{ierfc} \frac{2nL+x}{2\sqrt{\alpha t}} + \text{ierfc} \frac{2(n+1)L-x}{2\sqrt{\alpha t}} \right\} \quad (7)$$

where the constants are defined as previously. An alternative form of (7) is

$$w = \frac{aHt}{\rho cL} + \frac{aHL}{k} \left\{ \frac{2L(L-3x)+3x^2}{6L^2} - \frac{2}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} e^{-\alpha\left(\frac{n^2\pi^2}{L^2}\right) t} \cos n\pi \left(\frac{L-x}{L}\right) \right\} \quad (8)$$

which shows the behavior of $w$ at large values of $t$ more easily. For very short times, at $x = 0$, $w$ has approximately the same values as for the semi-infinite case, so that the variation of $Q_p$ with $t_p$ should be the same. However, at times exceeding

$$\frac{L^2}{2\alpha}$$

the variation of $w$ with $t$ is approximately linear, since the last term in (8) is negligible. Under these conditions we may write

$$w \approx \frac{aHt}{cL} + \frac{aHL}{k} \frac{2L^2}{6L^2}$$

$$\approx \frac{aH}{k} \left[\frac{\alpha t}{L} + \frac{L}{3}\right] \tag{9}$$

Thus for $x = 0$, the maximum temperature is again attained at $t = t_p$, and

$$Q_p = Ht_p$$

$$= \frac{w_m k}{a} \quad \frac{3Lt_p}{3\alpha t_p + L^2} \tag{10}$$

For depths below the surface the maximum temperature is attained after $t_p$. Since the pulse is cut off before this maximum has been reached, it is again necessary to form a solution for a time $t > t_p$, or

$$w = \frac{2aH}{k} \left\{ \sqrt{\alpha t} \sum_{n=o}^{\infty} \left[ \operatorname{ierfc} \frac{2nL+x}{2\sqrt{\alpha t}} + \operatorname{ierfc} \frac{2(n+1)L-x}{2\sqrt{\alpha t}} \right] \right.$$

$$\left. - \sqrt{\alpha(t-t_p)} \sum_{n=o}^{\infty} \left[ \operatorname{ierfc} \frac{2nL+x}{2\sqrt{\alpha(t-t_p)}} + \operatorname{ierfc} \frac{2(n+1)L-x}{2\sqrt{\alpha(t-t_p)}} \right] \right\} \tag{11}$$

This can be written as

$$w_m = \frac{2aH}{k} \{A - B\} \tag{12}$$

Numerical evaluation of (11) to obtain the variation of w with t, and thus determine $w_m$, would be extremely tedious because of the two series expressions. An alternative solution was to use an electronic analog computer to determine the values of $t_m$ and $w_m$. The computer was set up to simulate an insulated slab and a constant input voltage was used to produce the pulses of irradiation. The maximum voltage produced at a given point is

$$V_m = \frac{2aH}{k} \{A - B\} \tag{13}$$

in analogy with (12). This can be converted to reproduce the bracketed expression on the computer if we use a voltage

$$V_H = \frac{HL}{kN} \tag{14}$$

where N is the number of amplifiers used in the computer set-up. Then

$$V_m = \frac{2aN}{L} V_H \{A - B\} \tag{15}$$

We evaluated {A - B} for a number of pulse lengths, setting computer time equal to real time. Using equation (12) then

$$H = \frac{kw_m}{2a} \frac{1}{\{A-B\}} \tag{16}$$

and

$$Q_p = H t_p \tag{17}$$

so that

$$Q_p = \frac{kw_m}{2a} \frac{t_p}{\{A-B\}} \tag{18}$$

Equations (10) and (18) were used to evaluate the $Q_p$ variation with pulse length, again choosing the conditions $w_m = 500°C$, $a = 1$, $k = .001$ cal $cm^{-1}$ $sec^{-1}$ $°C^{-1}$, and $\alpha = .001$ $cm^2$ $sec^{-1}$. Here, however, we also had the condition $L = 0.1$ cm. These curves are plotted in Fig. 2, where curve IS(1) shows the variation of $Q_p$ at $x = 0$ and IS(2) shows the variation at $x = .05$ cm.

Curve IS(1) is the same as SI(1) until about 3 seconds pulse length; beyond that it approaches an asymptotic value due to its insulated condition. Curve IS(2) lies below SI(2) at all points, since the insulated slab loses no heat. Actually, however, we have found that the computer-simulated insulated slab does suffer from losses equivalent to slight cooling, so that the curve increases at both long and short times. The truly insulated slab of condition IS(2) should approach IS(1) at long times, since all the temperatures within the slab approach one another. At very short pulse lengths the true curve should also lie below that plotted. These probable curves are shown as dashed lines in Fig. 2. It is significant that curve IS(2), representing the mid-point of a thin (0.1) slab, slightly cooled, reaching 500°C, is reasonably flat from 0.1 to 10 seconds, and obeys the reciprocity relation even better than the same depth in a semi-infinite medium.

## IV. EXPERIMENTAL RESULTS

1. One of the first materials investigated in regard to the variation of its thermal damage with exposure time was a coated paper often used in this laboratory to record the intense images from our thermal radiation sources. This paper is actually made to serve as a liquid-vesicant detector in chemical warfare defense, and consists of a heavy (.010 inches) paper which is coated with a thin layer (approximately .0005 inches) of olive

green paint. The coated paper is designated as M6A1, and the paint as M5, Type 2, made by the Hilton-Davis Chemical Company, Cincinnati, Ohio, according to specification JAN-P-274. The paint apparently contains a red dye which is insoluble in the paint vehicle at normal room temperatures, but becomes soluble when liquid vesicants come into contact with it, or when the temperature is raised beyond some critical level. The red dye then becomes quite visible, and when exposed to beams of thermal radiation, it first turns red in an area corresponding to the shape of the beam. If the exposure produces still higher temperatures, the dye is separately deposited in a glossy melt. Beyond this the paper begins to char and burn. Each of these phenomena can be used as a criterion for studying reciprocity.

In Fig. 3, curve V shows how the exposure level at which the glossy melt appears varies from 0.1 to 10.0 seconds. The determination of this level is approximate, since it was obtained from a number of vesicant papers exposed to the image of the carbon-arc thermal source and mounted on a display chart showing a $Q_p$ vs. log $t_p$ variation. By matching similar surface appearances, approximate curves for the threshold phenomena were drawn directly on the chart. To obtain this data, the carbon arc was maintained at a given irradiance level while the pulse time was varied.

Curve V shows that for long times the exposure level becomes nearly constant with $t_p$, but that for short exposures, the required exposure decreases. The accuracy of the method is not sufficient to indicate whether the slope of the curve is significantly different from that for the front surface threshold of the insulated slab. This paint has a high absorptance in the ultra-violet and the visible regions of the spectrum, which makes it quite sensitive to radiation from the carbon-arc source.

Whether the reactions involved are time-dependent is not known to this writer, but the observed data are not inconsistent with a pure threshold temperature explanation which corresponds to the case of the insulated slab described previously. The very shallow depth of the paint layer, which is apparent in microscopic observation of the exposed papers, would also lead to this conclusion. A curve which shows the front surface slope for a semi-infinite medium is plotted in Fig. 3 for comparison.

2. A second phenomenon which has been of special significance in thermal radiation studies is the occurrence of disabling skin burns. In this laboratory such a burn (2+ mild) has been defined[6] as an area on a Chester-White pig where surgical examination 18-24 hours after exposure reveals that the epidermal injury permits easy separation of the epidermis from the dermis, and the latter is not appreciably damaged. Although the greatest interest in this laboratory has been in establishing levels of protection from such burns behind material assemblies, we have also accumulated several sets of data on the occurrence of burns on bare pig skin, where the severity is 2+M or more severe. Four such levels are indicated on Fig. 4, giving curve B-QM, which represents the exposure required to produce 50% 2+M burns in a set of exposures of white pig skin to the QM solar furnace beam. At exposure times of 0.5 and 1.0 seconds, the effective exposure for 50% burns was determined using statistical methods[6], but at 0.11 and 0.17 second exposures, the transition from no-burns to 100% burns was so abrupt that the mid-point of the small interval between these levels was taken to be the 50% level.

These burn data extend only over one order of magnitude in pulse duration, and indicate strongly that a minimum in effective exposure level

exists in the vicinity of 0.1 seconds. Previous studies[10,11] of such burns have indicated that the skin absorbs heat in a manner similar to that of a semi-infinite medium with the following average thermal properties:

$$k = .0018 \text{ cal cm}^{-1} \text{ sec}^{-1} (°C)^{-1}$$

$$\rho = 1.20 \text{ gm cm}^{3}$$

$$c = 0.86 \text{ cal gm}^{-1} (°C)^{-1}$$

$$\alpha = .0017 \text{ cm}^{2} \text{ sec}^{-1}$$

Since the average thickness of the epidermis is approximately 0.008 cm, it is interesting to compare the exposures needed to achieve a fixed maximum temperature at that depth below the surface. Using the relation (5), with $x/2\sqrt{\alpha} = 0.1$ and an absorptance value of $a = 0.5$, the curve of $Q_p$ versus $t_p$ was calculated so that it gave the same value of $Q_p$ as the experimental burn data at $t_p = 1.0$ second. At this pulse length a maximum temperature rise of 70 °C was required; curve SI-.008 thus produced and plotted in Fig. 4 then represents the exposure levels required to produce this maximum temperature at .008 cm beneath the surface of simulated skin at various thermal pulse lengths. This curve also approaches a minimum exposure, but in the region of $t_p = 0.01$ second.

It is evident that the slopes of curves B-QM and SI-.008 are not at all similar. This attests to the impossibility of explaining the burn phenomena, which are governed by a reaction rate strongly dependent upon temperature[11], in terms of a maximum temperature. A more useful criterion might be the integral of the temperature-time curve above some threshold value. Nevertheless, the position of the minimum in curve SI-.008 certainly shows that the maximum temperature, if used as a criterion, must occur

deeper within the skin in order to shift it to longer pulse durations.

Further experimental evidence for such burns can be obtained from the cumulative data of the University of Rochester[10], shown as curve B-UR in Fig. 4. These data (the averages of which are plotted in Fig. 4) cover two orders of magnitude, from 0.3 to 30 seconds. No evidence of a minimum is seen for the averages, although single experiments have indicated that it exists[12]. The slopes of the two experimental burn curves are essentially the same in the region where they overlap. The exposure levels of the curves are different, presumably because of the spectral differences between the solar furnace and a carbon arc source, as found in this laboratory[6].

Fig. 4 also shows two curves which portray the critical exposure to produce a temperature of 70°C at the surface of a solid with the "skin" diffusivity of .0017 $cm^2$ $sec^{-1}$ $(°C)^{-1}$ and an absorptivity of 0.5, both for the semi-infinite solid and for a slab of 0.1 cm thickness. These are labeled curves SI-0 and IS-0, respectively. From the calculated curves it is possible to construct a curve for attainment of 70°C temperature maximum at 0.008 cm beneath the surface of a 0.1 cm slab; such a curve would approach curve SI-.008 at short pulse lengths, and curve IS-0 at large pulse lengths.

3. The behavior of thermally irradiated fabric is also particularly interesting over a wide range of exposure durations. In order to include this behavior it was necessary to select a specific fabric for study: a cotton poplin Army fabric, 5 $oz/yd^2$, green shade 116. This material was exposed to the solar furnace beam while in contact with a transite block at room temperature. Samples exposed at a given pulse length were examined carefully and the exposure levels at which selected criteria began to

appear were noted. The criteria used were (a) discoloration of the exposed surface, apparently due to a tar deposit, (b) charring, caused by complete decomposition of cellulose, (c) sustained flaming, with burning through the fabric. The minimum levels for each of these criteria are shown in Fig. 5. In addition, the occurrence of ignition during exposure was also noted and used as a separate criterion, also plotted in Fig. 5.

The slopes of the curves in Fig. 5 all seem to be quite similar, but in view of the empirical nature of such data this cannot be taken too literally. The curves for burning-through and ignition seem to merge at long pulse lengths, as would be expected when the heating takes place more slowly. For exposures longer than 10 seconds the curve for ignition has a slope which is much higher than that observed or calculated for the criteria used with other materials. This slope approaches that of a line of constant irradiance (H) drawn on the $Q_p$ versus $t_p$ chart. The H value at which the criterion becomes independent of $Q_p$ is approximately 3.4 cal $cm^{-2}$ $sec^{-1}$. The meaning of this behavior is that ignition depends only upon H at long times and not upon the length of the exposure. Since prolonged ignition produces burning through the fabric, it is not surprising that the curve for the latter criterion becomes identical with that for ignition.

At short times the ignition curve coincides with the curve for charring. This, too, is reasonable since the processes must take place rapidly and ignition will thus follow charring extremely closely.

4. In order to include more diverse materials within this study, 1/4" thick samples of rock maple were also exposed to the image produced by the solar furnace. These samples were cut from a 1" x 1" square rod on a circular saw, and the faces were exposed without further smoothing.

Each sample was held during exposure in the center aperture of a water-cooled plate. As before, samples were exposed with a variety of irradiation levels and pulse lengths.

A two-dimensional array of the exposed samples was made, showing exposure levels as ordinates and pulse durations as abscissae. From this array, data could be extracted to draw smooth curves for the charring of the wood samples. The two criteria used were the beginning of charring on the exposed face, and complete charring of the face, both visually determined from the array of samples. The curves for both of these criteria are shown in Fig. 6. The slope of both curves suggest that sub-surface effects do not control the appearance of charring appreciably.

5. Samples for radiant exposure were also made from a 1" diameter nylon rod. These were cut on an electric hack saw to have an average thickness of 1/8". The nylon has a high reflectance and is also partially transparent, so that higher exposure levels are required in order to produce suitable changes in the material. These were achieved by using a second ellipsoidal mirror in tandem with the carbon arc image source, such that the far focus of each mirror was at the same point. Thus an image of the carbon arc crater was formed at the near focus, of approximately the same dimensions as the source itself. This image was quite small, and had a very large angle of convergence. By the use of attenuating screens in the common focal point of the system the irradiance could be varied, and the shutter was used to cut the beam near this point as in normal carbon arc operation.

A large number of samples was exposed in this way at irradiances up to 80 cal $cm^{-2}$ $sec^{-1}$ and over a pulse duration range of 0.1 to 20

seconds. The exposure level at which melting just began on the nylon surface was determined by comparing these samples at the various pulse lengths. A second level at which the melt had the approximate dimensions of the image area was also determined from the assembled data. Both of these empirical curves are plotted in Fig. 6. No effect other than the formation of a brown melt was visible. The diathermancy of the nylon samples caused heating of the samples all the way through; however, the curves indicate that the behavior is still somewhat like that of a surface phenomenon.

## V. DISCUSSION OF RESULTS

Studying the response of a material to thermal radiation in terms of its departure from reciprocity has several elements of the empirical, or heuristic, approach, with attendant disadvantages. The criterion of response is in most cases a change in the state of the material, visually determined. Quantitative measures of such changes are preferable, and would undoubtedly yield results with less "spread" in the determination of a critical exposure. However, the development of a quantitative method for each material would grow into a series of separate research problems, and while useful in themselves, these would tend to obscure the general view of material behavior which is desired here.

A more serious criticism of the method pertains to the seemingly arbitrary selection of the criterion. For materials whose use demands that certain changes in state are not tolerable, the selection was not really arbitrary. For others, an easily identifiable change was selected, and it is of course possible to choose other criteria. Such choices might be pertinent and might lead to varying conclusions. The variety of materials

and criteria of damage used in these discussions is used only to portray a pattern of behavior, and not to yield information about specific physical or chemical processes.

The form we have used for plotting the data may seem cumbersome, particularly for the calculated curves based on maximum temperatures. However, the basic philosophy of irradiation studies rests upon the calorimetry concept, or conservation of heat energy. From this point of view the most fundamental parameter in a study of behavior under irradiation is the total energy absorbed, or the exposure. A true calorimeter would obey the reciprocity relation completely, giving the same response at a particular exposure value, regardless of the duration of irradiation. Actual systems may be constructed which do measure this exposure over a range of pulse durations, such as our copper disk calorimeters and some photochemical solutions. This makes the critical exposure an independently measurable quantity, and materials can thus be classified according to their deviation from the ideal calorimeter response. Such deviations can obviously be produced by a variety of causes, but these are probably not as important in the overall view as the amount and direction of the deviations from reciprocity. These could be plotted differently without making significant changes in interpretation. For example, we could plot critical exposure ($Q_p$) versus irradiance (H), critical exposure ($Q_p$) versus reciprocal irradiance ($t_p/Q_p$), or critical irradiance (H) versus pulse duration ($t_p$). In practice, none of these have yielded any new information about the processes involved. Furthermore, the plot of exposure ($Q_p$) versus pulse duration ($t_p$) is the most appropriate in the experimental sense, for the reasons discussed above. Criticism of the logarithmic scales has often been

voiced, but we have found no better way to portray the changes encountered over several orders of magnitude in time, and usually about two orders of magnitude in exposure. Furthermore, the use of a linear pulse time scale tempts us to the "zero extrapolation", a process which is not at all justifiable unless reciprocity is obeyed absolutely. The concept of an instantaneous heat pulse (delta function) is a useful mathematical abstraction, but it cannot be realized through graphical extrapolation of experimental results.

Obviously, there is no "typical" response of a material to thermal radiation. Such phenomena, however, often have curves which show similar regions of variation in $Q_p$ with $t_p$. At long times the slope on the log $Q_p$ versus log $t_p$ chart is positive, varying between zero and one, if the chart scales are the same. Zero slope, of course, represents constant $Q_p$ values, or perfect reciprocity, while a slope of one represents constant H values, or dependence only on the irradiance. It is difficult to image a response which demands either a decreasing critical exposure at very long times, or which requires an increasing irradiance at such long times. Thus for long pulses all of the materials we have examined stay within the positive slope region, between zero and one.

Many phenomena also exhibit a region where the critical exposure goes through a minimum. In general this behavior is related to a subsurface condition which must be present before the required criterion appears. Curves which exhibit such a minimum probably go over to a negative slope at still shorter pulse lengths, although the data are too scanty to establish this. The nature of these phenomena indicates a definite need for very high-intensity, short-pulse sources of thermal

radiation. Ideally, sources which yield 10 calories per square centimeter in pulses of a few milliseconds are desirable, but this would require an irradiance greater than 1000 calories $cm^{-2}$ $sec^{-1}$.

This negative slope is also clearly shown in the calculated curves for sub-surface attainment of a given temperature maximum. This seemingly odd result is simply due to the fact that although the pulse time may approach zero (the abstraction of the instantaneous heat pulse), so that the irradiance approaches an infinite value, nevertheless both the critical exposure and the time required for registration of a sub-surface temperature maximum remain finite. For a medium of known thickness and thermal properties, these parameters are easily calculable[9].

In the experimental (or empirical) case we do not expect materials to behave like these calculated curves for short pulses. The requirement that H increase even more rapidly as $t_p$ decreases will undoubtedly bring in new changes of state which destroy the constancy of exposure conditions. Thermal shock and subsequent ejection of material from the surface would make it impossible to carry these particular damage curves into the microsecond region. However, for pulses of the order of milliseconds it is certainly possible to continue using the damage criteria we have defined. At shorter pulse lengths new criteria would probably be need to be set up.

The most useful result which could be derived from this sort of study of thermal damage is an understanding of the basic phenomena taking place in the material. This would make it possible to predict the behavior of similar materials and to extend the applicability of experimental data to situations and conditions not easily duplicated or simulated in the laboratory. Unfortunately, this result can seldom be realized; the best

we can do is to obtain a qualitative understanding of the phenomena and the effects which variations of the parameters will bring about.

In Fig. 7 are plotted several curves which show the behavior observed in the study of thermal damage. They represent idealized relationships which might be limiting cases for specific materials. Also shown are schematic indicators for various mechanisms which affect the slopes of the curves. Fig. 7 illustrates the main point of this paper, namely that the behavior of some specific phenomenon must be placed within the framework of the known behavior in idealized media in order to classify and understand its dependence of $Q_p$ on $t_p$. While this procedure may not yield basic information about the chemical and physical changes which take place, it can help us compare the material with one whose reactions are more completely understood. It can also aid us in planning the alteration of the material or in selecting its area of best employment.

All of the phenomena studied experimentally in this survey developed irreversible changes, while the curves calculated in III, B, simply dealt with a maximum temperature achievement, a reversible condition. These are not as divergent as they might seem, but some of the irreversible phenomena we classify are related to the achievement of a specific temperature and others are produced by temperature-dependent reactions. The relationship is seldom a linear one; volatile products and latent heat are usually involved. These factors tend to offset one another, so that it is not possible to give any general rules regarding the effect of a departure from reversibility. Since thermal damage is presumed to imply a permanent change in the material, reversibility is of importance only in interpretation of the results or in the simulation of other phenomena in laboratory experiments.

In connection with the simulation problem, which is of particular importance to nuclear weapons thermal effects, more attention might be given to the "reciprocity" analysis presented here. If a certain irreversible criterion, such as burning of skin, can be classified in regard to its $Q_p$ versus $t_p$ behavior, then any reproducible phenomenon which can be forced to yield the same curve, under the same environmental conditions, could be considered as a suitable simulant. It is not necessary that it actually simulate the thermal or chemical properties of the material, and it is preferable that its response be reversible so that it could be used repeatedly. The approach is frankly empirical, and it would not aid in understanding the fundamental processes taking place. However, it would provide a workable laboratory simulator, with different criteria being used for the various environments employed.

From Fig. 4 it can be confirmed that the bare skin burn data do not conform to the attainment of a temperature maximum on or beneath the surface of a solid. However, it may be possible to find a value of the parameter $x/2\sqrt{\alpha}$, a maximum temperature, and a slab thickness such that the curves of $Q_p$ vs. $t_p$ would nearly coincide. This material could have higher values of $x$, $\alpha$ and $L$ and would not need to be similar to skin at all. In general, a simulant would be more useful in experiments with clothed skin; for this new values of the parameters would need to be derived and a heat acceptance rate matching that of skin would be achieved. Such an approach avoids the necessity of achieving exact simulation of the thermal properties and subsequent interpretation of temperature data in terms of thermal damage.

## VI. CONCLUSIONS

The extraction of definite conclusions from a survey of this type is difficult. The main purpose of the work was to point out similarities in behavior of diverse materials, to aid in an understanding of this behavior in terms of theoretical relationships and to indicate the usefulness of the empirical approach.

The first objective was achieved by using a common method of data presentation for existing data and for some empirical data from five experimental studies. The dominant factors in the achievement of a damage criterion were in most cases identifiable.

The second aim was achieved by using attainment of a given temperature maximum, either on the surface or within a solid material, as a conceptual guide for curves of exposure versus pulse duration. The generality of these curves, even though the criterion is artificial, gave us a very useful framework for examination of the experimental data. The existence of minima in these curves, which approximate reciprocity over large ranges of pulse length, was shown to be a function of effective damage depth and thermal properties. It is hoped that this result will help put "reciprocity" in its proper perspective, although it is not intended as an explanation of any of the experimental data.

The third objective concerns the need for simulation of thermal damage effects in laboratory experiments. Without suggesting any specific materials, it was pointed out that coincident curves of $Q_p$ versus $t_p$ would signal complete success of the simulation effort, regardless of the processes involved in either the simulant or the simulated material. The only disadvantage is that good data on both materials are required in order

to establish this coincidence of curves. However, this would be true for any method of simulation, no matter how exact it was expected to be.

A final conclusion, which perhaps need not be explicitly stated, is that no thermal damage criterion should ordinarily be expected to show reciprocity. If it does seem to give such a relationship it is probably due to the restricted range of pulse lengths studied, or to a dominant photochemical reaction.

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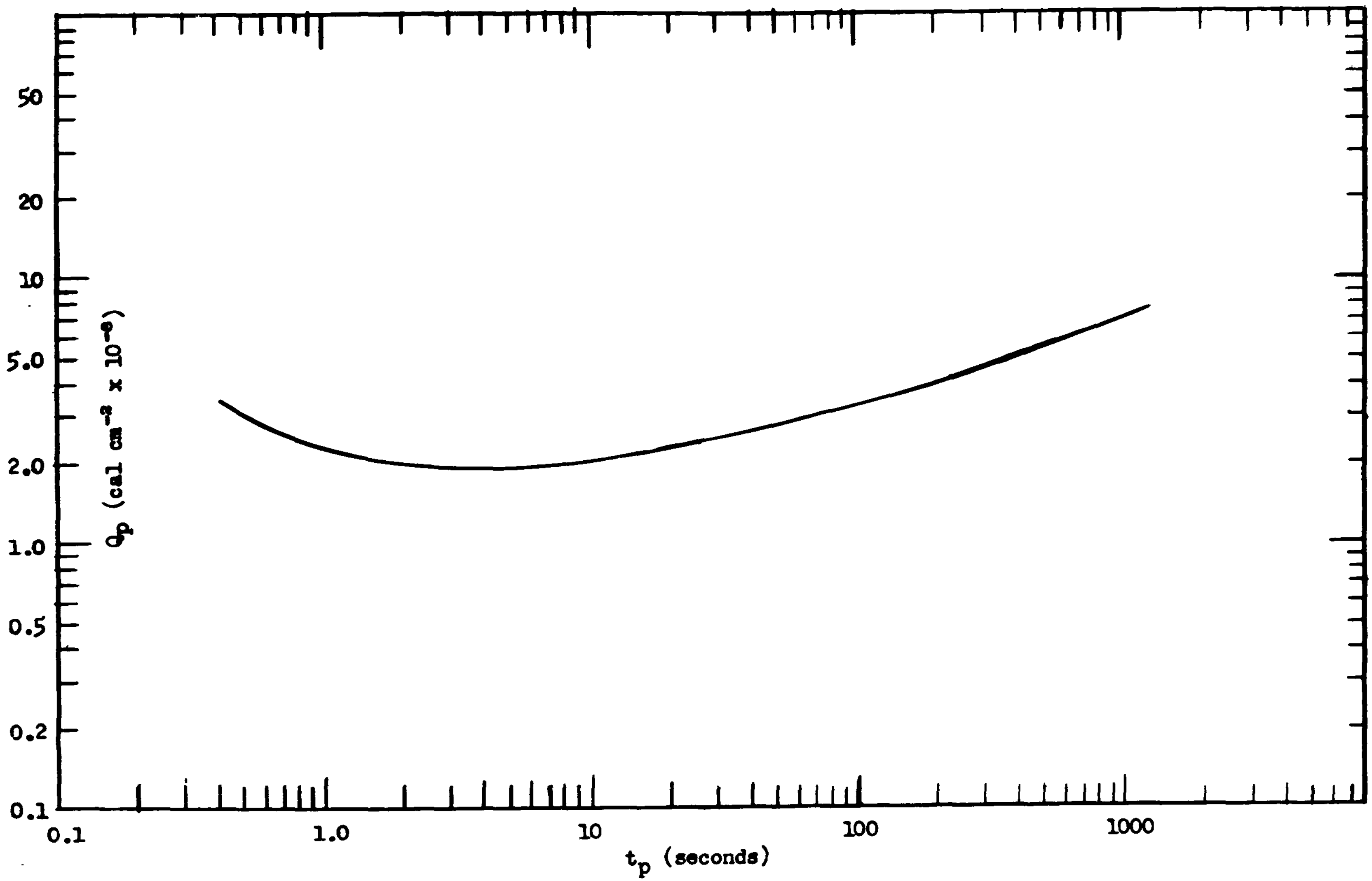


Figure 1. Critical Exposure for Constant Density in Photographic Emulsion

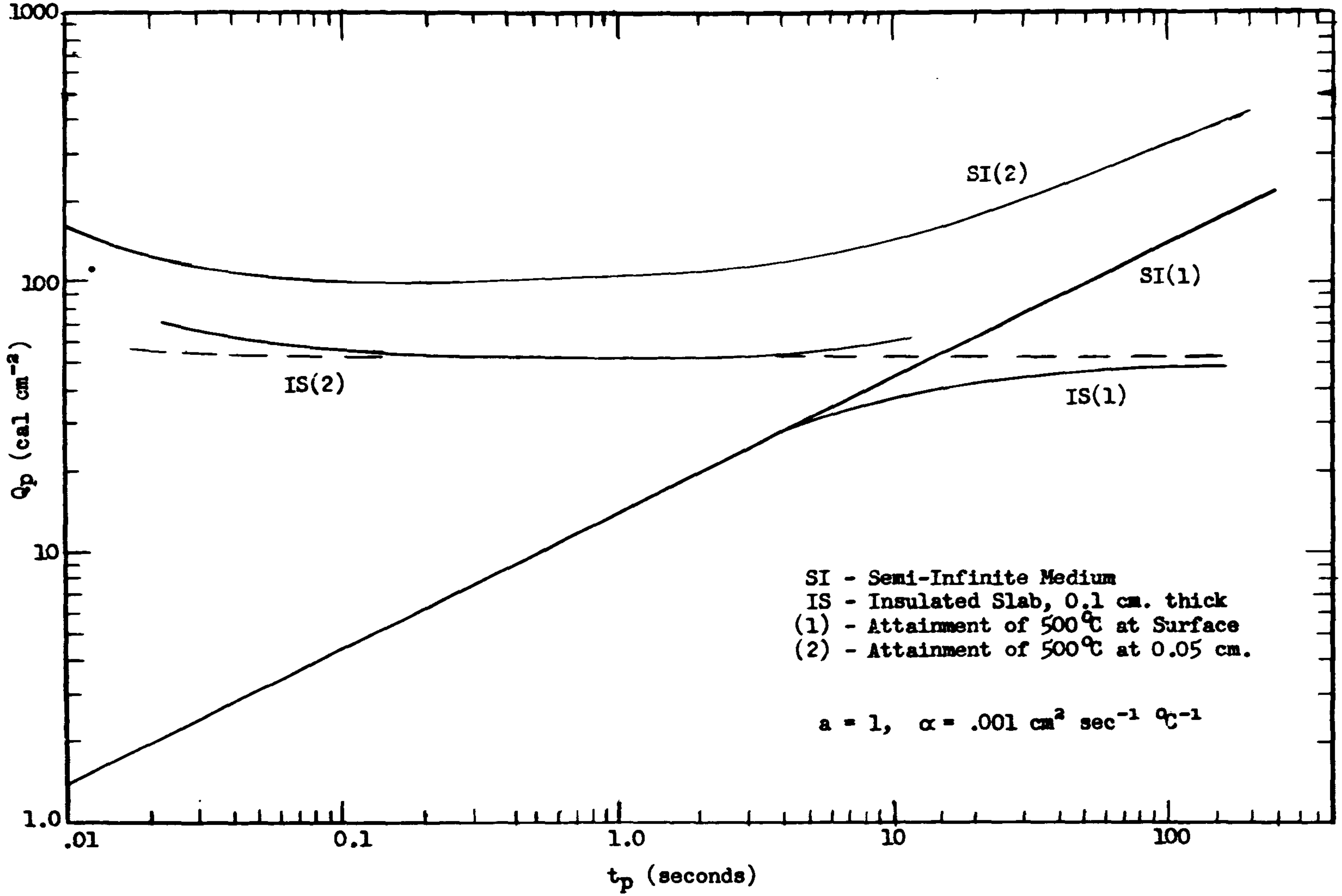


Figure 2. Critical Expcsure for 500°C Maximum Temperature Rise

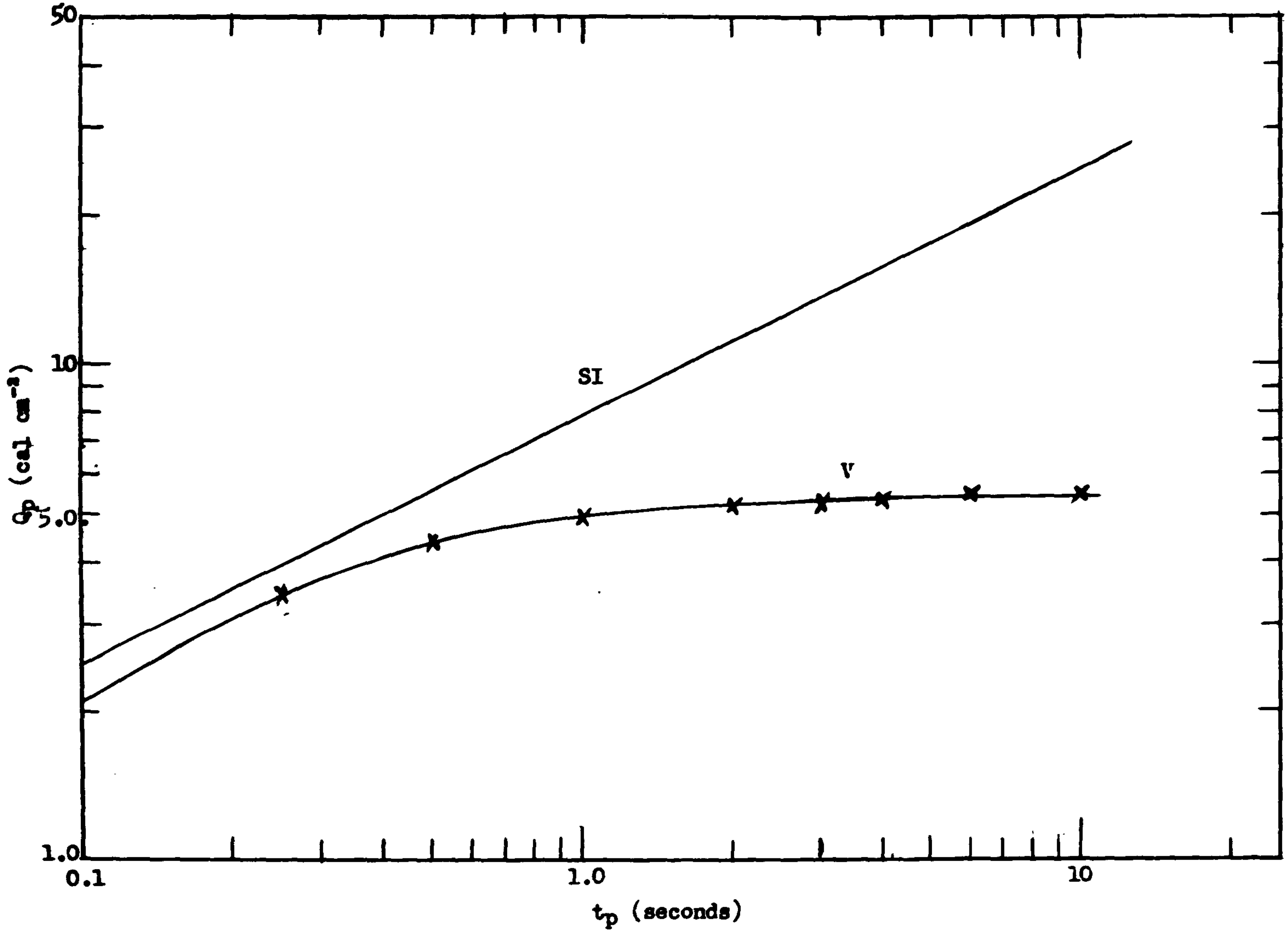


Figure 3. Critical Exposure for Melting of Dye on Vesicant Paper

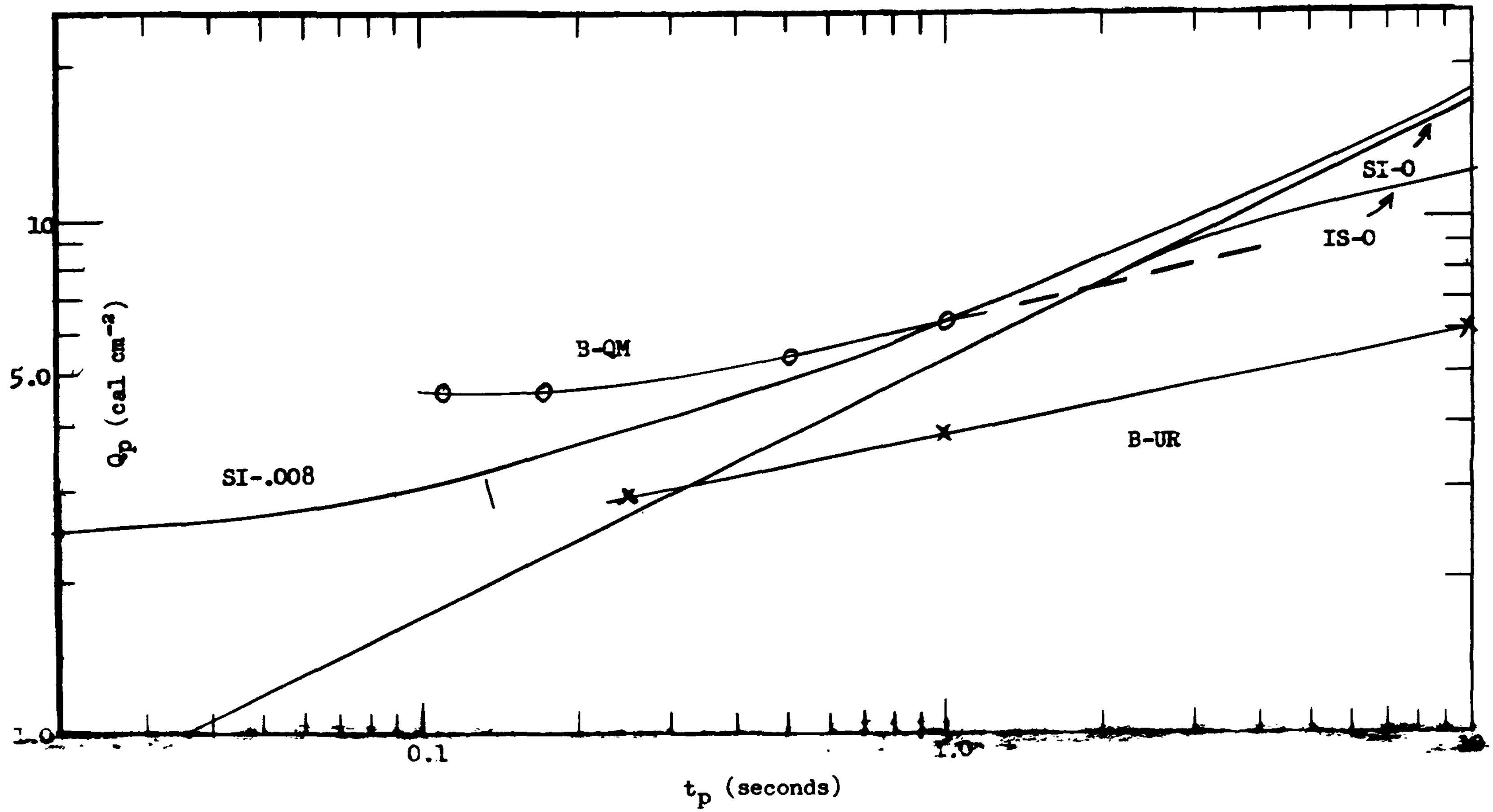


Figure 4. Critical Exposure for 2+ Burns as a Function of Pulse Length

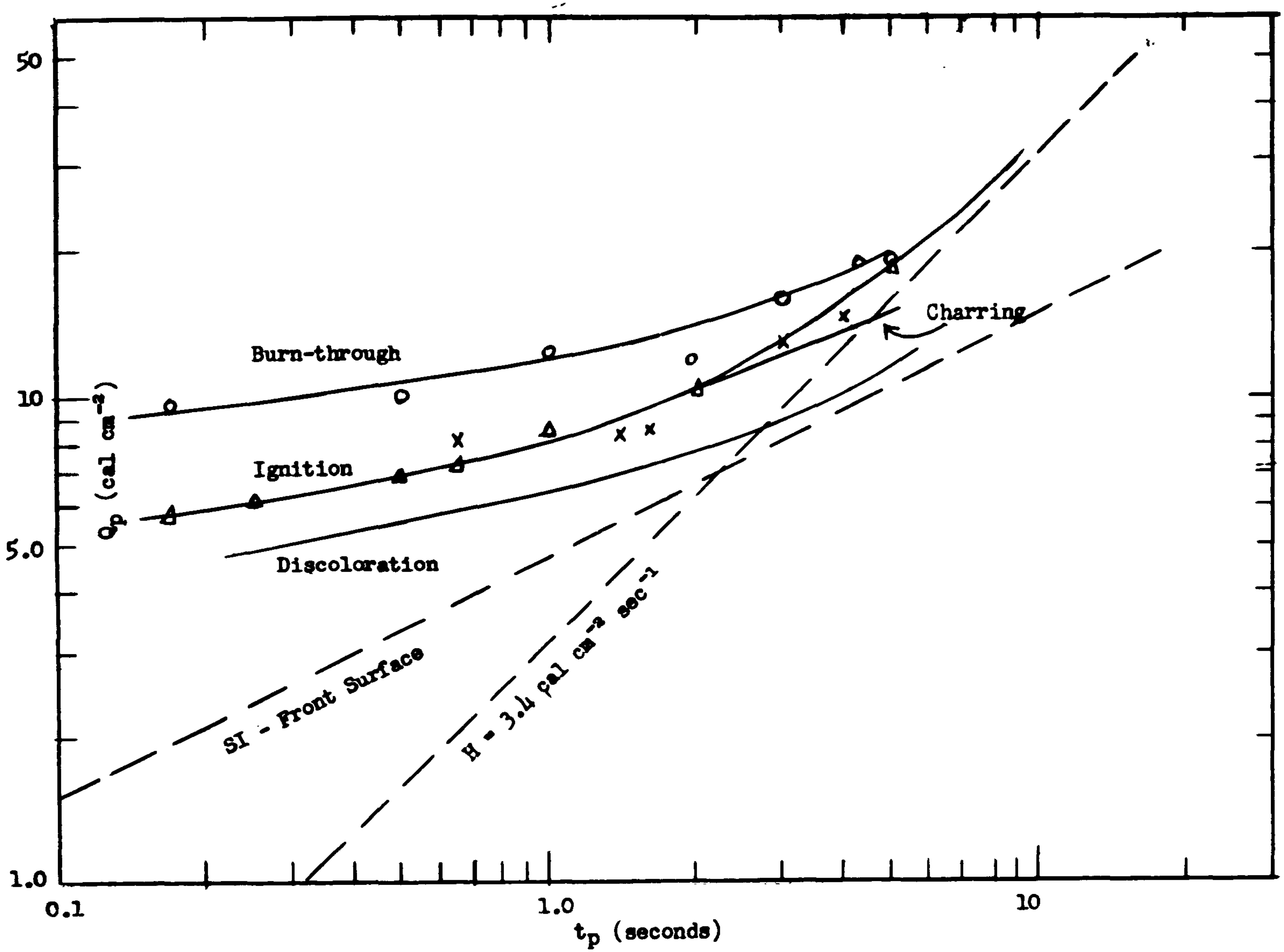


Figure 5. Critical Exposures for 5-ounce Poplin on Transite

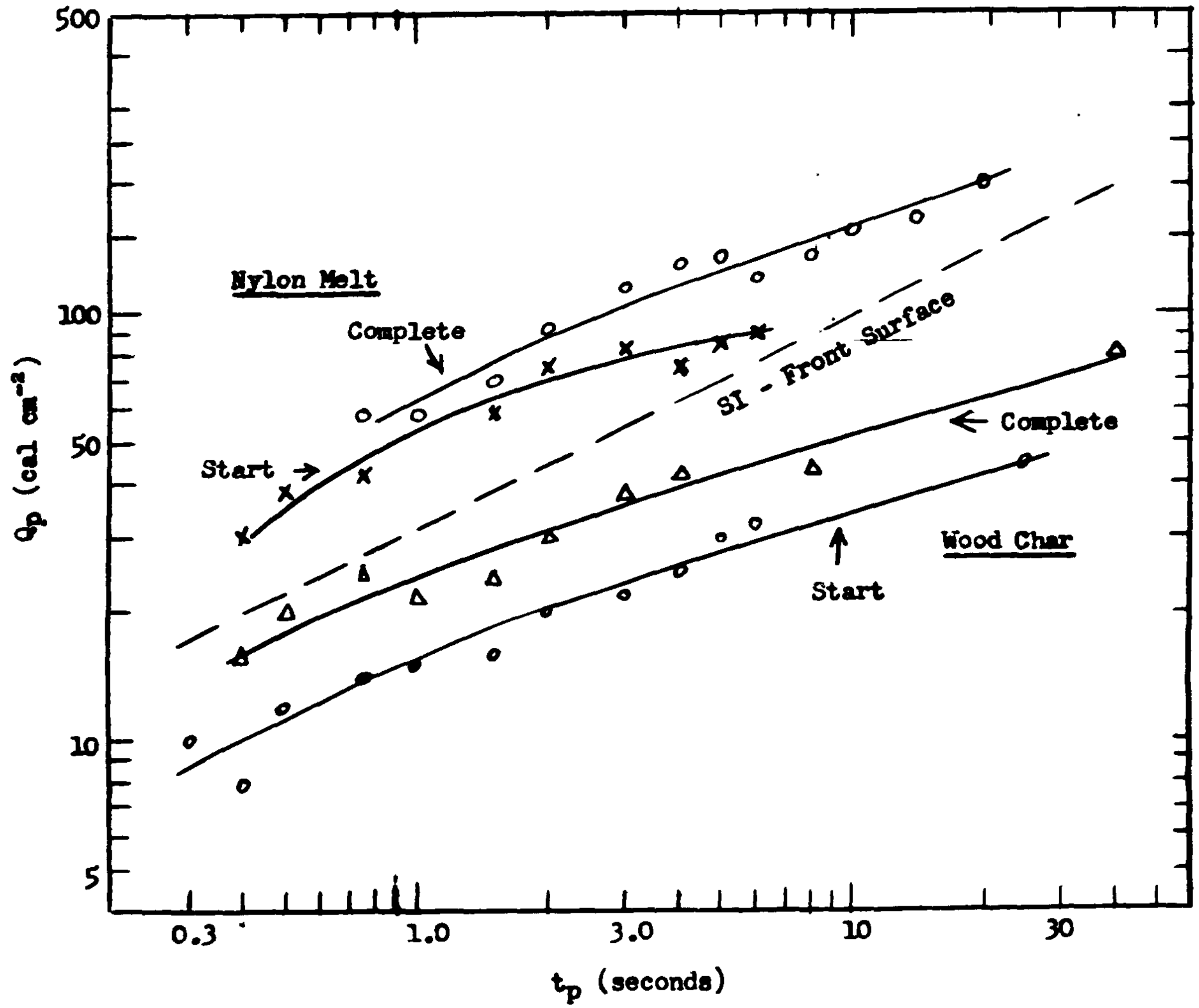


Figure 6. Critical Exposures for Nylon and Wood Slabs

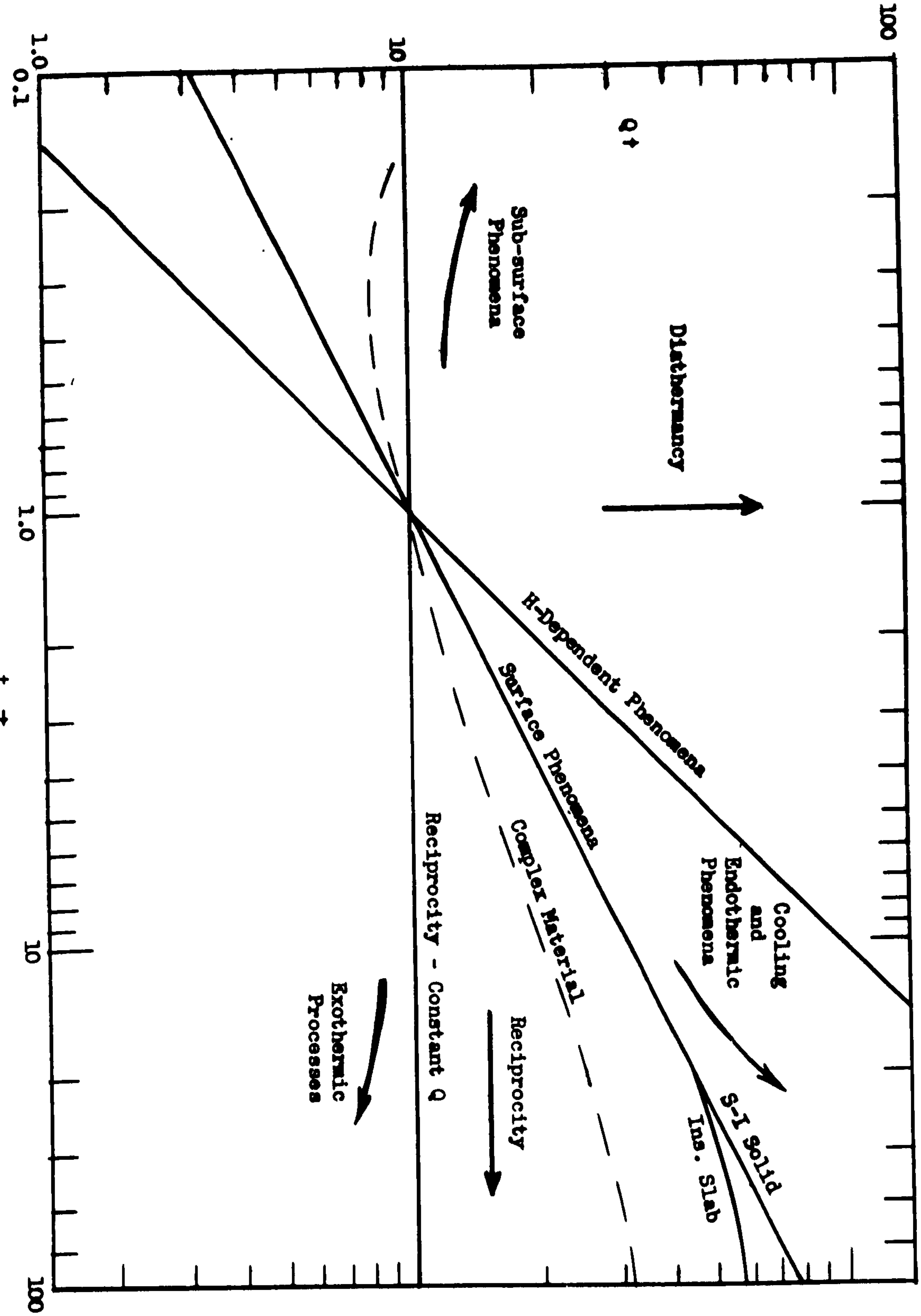


Figure 7. Idealised Forms of Exposure-Pulse Time Curves